

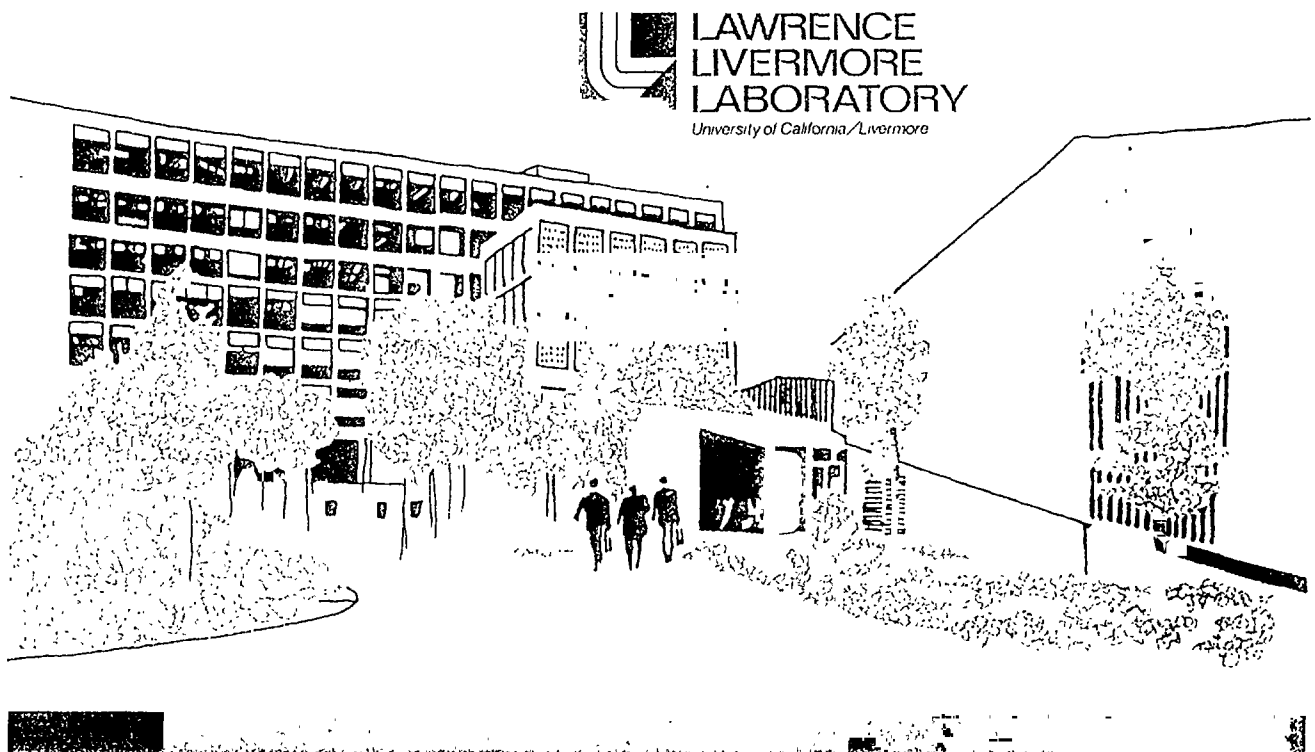
PLUTONIUM CONCENTRATIONS IN DIETARY AND INHALATION PATHWAYS AT BIKINI AND NEW YORK

W. L. Robison

V. E. Noshkin

September 27, 1976

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PLUTONIUM CONCENTRATIONS IN DIETARY AND INHALATION PATHWAYS AT BIKINI AND NEW YORK

Abstract

This report assesses the plutonium intake via inhalation and ingestion for residents of New York and residents of Bikini Atoll. Based on inhalation and ingestion intake, the plutonium transferred to urine would be roughly seven times greater at Bikini than at New York. This ratio compares with data reported to the authors showing urine samples from Bikini residents had about ten times the plutonium of urine samples from New York residents. The comparison of these ratios indicates that differences in the levels of intake in the New York and Bikini populations can account for the relative difference between the plutonium concentrations observed in urine samples from the two locations.

However, transfer coefficients and models presently accepted for inhaled

and ingested plutonium do not account for the absolute magnitude of the observed urine-sample concentrations. This discrepancy suggests inaccuracy in either the plutonium concentrations reported for urine samples or the models and transfer coefficients presently accepted for the metabolism of plutonium in the body.

The available urine data consist of results from very few samples, and more data is needed to completely verify the assessments of this paper. It is felt that Bikini Atoll may be the only global sources of data on humans where intake via ingestion is thought to contribute the major fraction of the plutonium body burden, and therefore Bikini is possibly the best source of data on transfer of biologically incorporated plutonium across the gut wall.

Introduction

Bikini Atoll was one of the two major locations for United States nuclear tests from 1946 through 1958. Rehabilitation of the atoll began in

1969-70. In 1970, analyses by the HASL* ERDA Laboratory of pooled urine

*Health and Safety Laboratory, New York, N.Y.

samples from Bikini Island showed the average concentration of $^{239,240}\text{Pu}$ to be 0.007 pCi/liter.¹ In 1971, the average concentration of $^{239,240}\text{Pu}$ in three urine samples from Bikini was 0.004 pCi/liter. But in 1974 the average in ten samples was a much higher 0.013 pCi/liter.¹ (No errors are attached to these concentrations reported in Ref. 1. The average concentration for any year was obtained by multiplying the plutonium concentration of each sample by its volume, summing these products, and dividing by the total volume.) In 1975 the average $^{239,240}\text{Pu}$ concentration in a pooled 9-liter urine sample from Bikini was reported as 0.011 pCi/liter.² This concentration is similar to the value found in 1974 but is ten times higher than the concentration of 0.001 pCi/liter reported for a 1975 representative population sample from New York.²

Because of the toxicity of plutonium and the belief that urine concentrations directly indicate plutonium burdens in the body, the 1970-75 increase of plutonium in Bikini urine samples and the much higher plutonium urine levels in Bikini samples, compared to New York samples, are cause for concern.

The purpose of this report is to compare data on Bikini and New York plutonium pathways to man. Although comparing the intake of plutonium via

various pathways does not lead directly to absolute plutonium concentrations in body tissues or excretions, it does show that a Bikini population is exposed to higher plutonium levels through dietary and inhalation pathways than a New York population.

We acknowledge that the excretion rates of plutonium and the quantities excreted may differ significantly, depending on routes of entry into the body, and that the assessment of these rates and quantities is further complicated by dissimilar physio-chemical forms of plutonium in the environment. We suggest that different concentrations of plutonium in corresponding pathways of New York and Bikini can account for the difference between urinary levels presently found in the two populations. However, the estimated intakes of plutonium at both New York and Bikini should give concentrations in urine very different than those reported to us.² This discrepancy suggests several interpretations.

If the concentrations found in pathways relate directly to the concentrations excreted, then the plutonium in Bikinians' urine will increase with time and will differ even more from concurrent New York samples as the Bikinians rely increasingly on dietary components from their atoll.

Pathway Analyses

PLUTONIUM CONCENTRATIONS IN THE INHALATION PATHWAY

The contribution from the inhalation pathway to plutonium concentration in urine, especially at Bikini, is very hard to quantify. In addition to the different activity levels from fallout in the air at Bikini and New York, resuspension processes that contribute airborne plutonium at New York and Bikini are very difficult to assess. Nevertheless, it is important to assess these processes in estimating the cumulative exposure to plutonium from inhalation. Bennett³ has recently concluded that plutonium from resuspended fallout at New York presently contributes 0.3% and will ultimately contribute intake equal to 0.5% of the intake that occurs during the original deposition of the fallout debris. Data of one of the authors⁴ show that the soil plutonium levels on Bikini Island are, on the average, higher than plutonium levels from fallout deposited in the United States.⁵

Visual observations indicate that the formation of dust clouds or the resuspension of surface materials by people or vehicles at Bikini is slight, even during very long dry spells. However, the available aerosol data show that some locations on Bikini Island have more plutonium

in the air than expected from world-wide fallout and more than encountered in New York City during comparable periods. These data indicate that resuspension does significantly contribute to inhalation of plutonium at Bikini. We will show in a following section that resuspension might also contribute to elevated plutonium levels in the ingestion pathway at Bikini.

Comparable data on plutonium concentrations in the air exist only for the periods of May 29 to June 2, 1970,⁶ and May 1972.⁷ During the 1970 sampling period, ^{239,240}Pu levels in surface air were determined at five locations on Bikini Island and ranged from 60 to 540 aCi/m³.⁶ The mean air concentration at four sites on the island of Eneu (in Bikini Atoll) was only 40 aCi/m³ during a comparable period.⁶ Plotted in Fig. 1 as a function of latitude are the average plutonium concentrations at ground level at air sampling stations of the HASL sampling network, taken during June 1970.⁸ The concentration-latitude profile is simply a continuous curve drawn through the available data points.

From this curve, the average fallout-plutonium concentration expected in ground-level air at the latitude of Bikini Island (11°, 37'

N) is approximately 32 aCi/m^3 . The mean of concentrations detected on the less contaminated island of Eneu agrees well with this expected concentration.^{4,9} However, on Bikini

Island the log normal median, the mean of measured concentrations (134 and 186 aCi/m^3 , respectively), and even the lowest concentration found on the entire island are well above

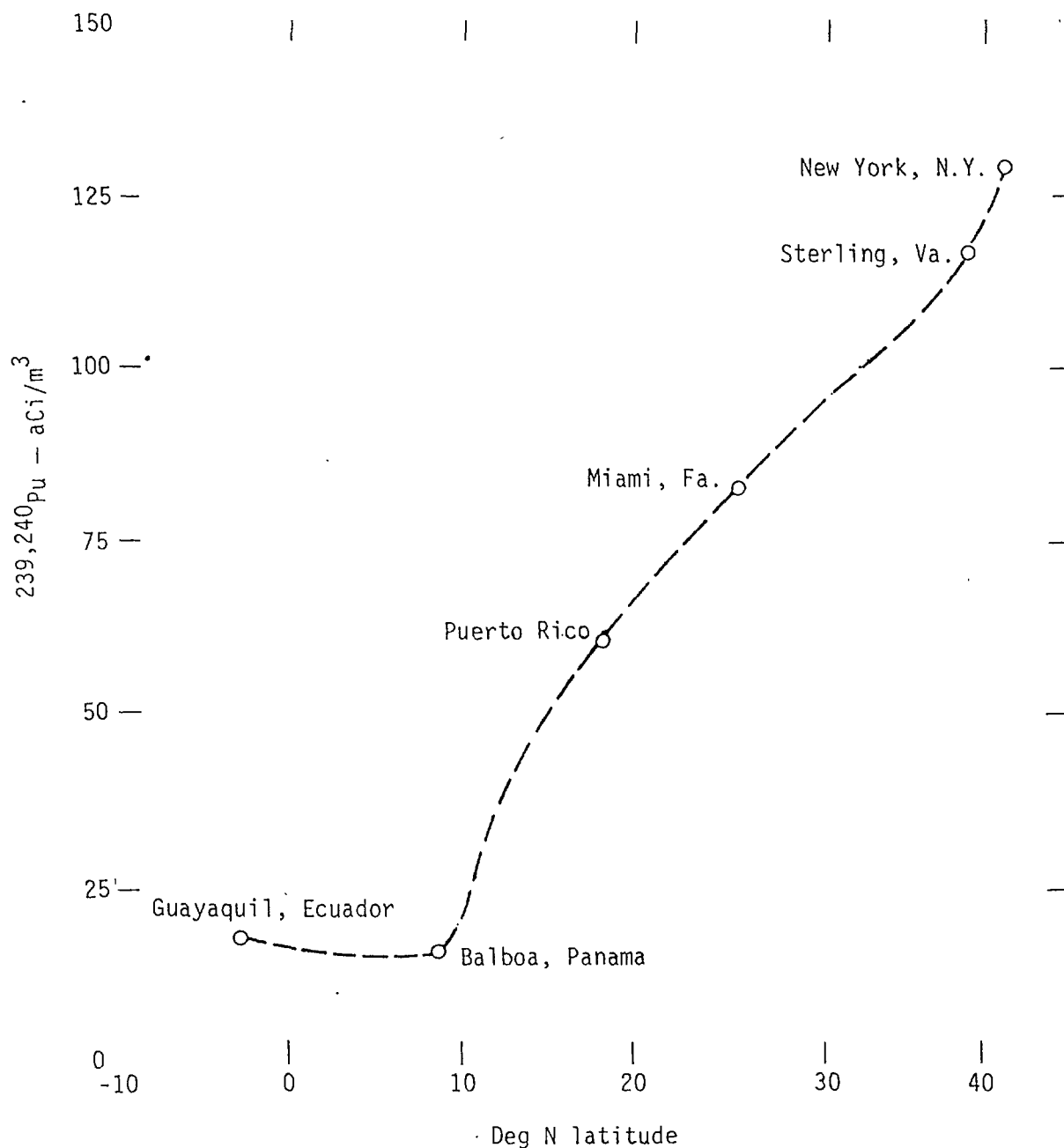


Fig. 1. Concentrations of $^{239,240}\text{Pu}$ in air at various latitudes, taken during June 1970, by the HASL sampling network.

the concentration expected from Fig. 1. Thus, there is little question that the plutonium levels in the air of Bikini Island around the first part of June 1970 were above fallout-plutonium background levels for that latitude. The only mechanisms that could cause such levels are resuspension processes on the island.

The median concentration at Bikini was very nearly equivalent to the mean air concentration at New York during this period in 1970. Remember that during 1970 the urine concentrations from Bikini and New York were also comparable. Except for water and possibly some fish, no indigenous material was consumed at Bikini during 1968 and 1969. From this comparison of New York and Bikini, the 1970 data strongly suggest a close correlation between plutonium levels in air and concentrations in urine.

In May, 1972, air samplers were again operated on Bikini Island at four different locations. The $^{239,240}\text{Pu}$ air concentration during this period ranged from less than 6 to 80 aCi/m^3 .⁷ The log normal median concentration over the island was 21 aCi/m^3 , and the mean concentration was 34 aCi/m^3 . In Fig. 2 the HASL air concentrations of plutonium during May 1972 are plotted as a function of latitude.⁸ A smooth continuous curve has again been drawn

through the data points. From this curve, the concentration of plutonium expected in air at the latitude of Bikini during May 1972 is approximately 16 aCi/m^3 . While the log normal median (21 aCi/m^3) of actual concentrations at Bikini is only slightly more than this value, the mean plutonium air concentration detected at Bikini during this period (34 aCi/m^3) was twice this expected value. In this same month, 37 aCi/m^3 were detected in the air at New York. Again we find a similarity between the levels of plutonium in Bikini and New York air, and the Bikini mean concentration is again higher than expected for this latitude from worldwide fallout concentrations.

The 1972 data on air concentration of plutonium at Bikini also showed a strong geographic correlation.⁷ The plutonium air concentrations in the samplers increased from northwest to southeast along the length of the island. Therefore, some regions of the island have higher plutonium air concentrations than others. The individual's inhalation exposure, then, must also depend on the time spent in specific regions of the island.

In summary, the data on air concentrations of plutonium at Bikini are rather sketchy, and we do not know how different the concentrations in large volumes of open air are from

those in an individual's immediate environment, resulting from resuspension. We can at least safely assume that the plutonium intake via the inhalation pathway on Bikini Island is comparable to that in New York. Also, the available data strongly suggest that over certain regions of the island the aeolian concentrations are significantly higher than in New York. If resuspension by human activity is also important, as it seems to be, there

is a much greater chance of higher exposure at Bikini through the inhalation pathway.

PLUTONIUM CONCENTRATIONS IN THE INGESTION PATHWAY

Drinking Water

The primary source of drinking and cooking water for Bikini inhabitants is unprocessed rain water obtained from cisterns attached to the newly constructed buildings along Lagoon Road. The cisterns collect water

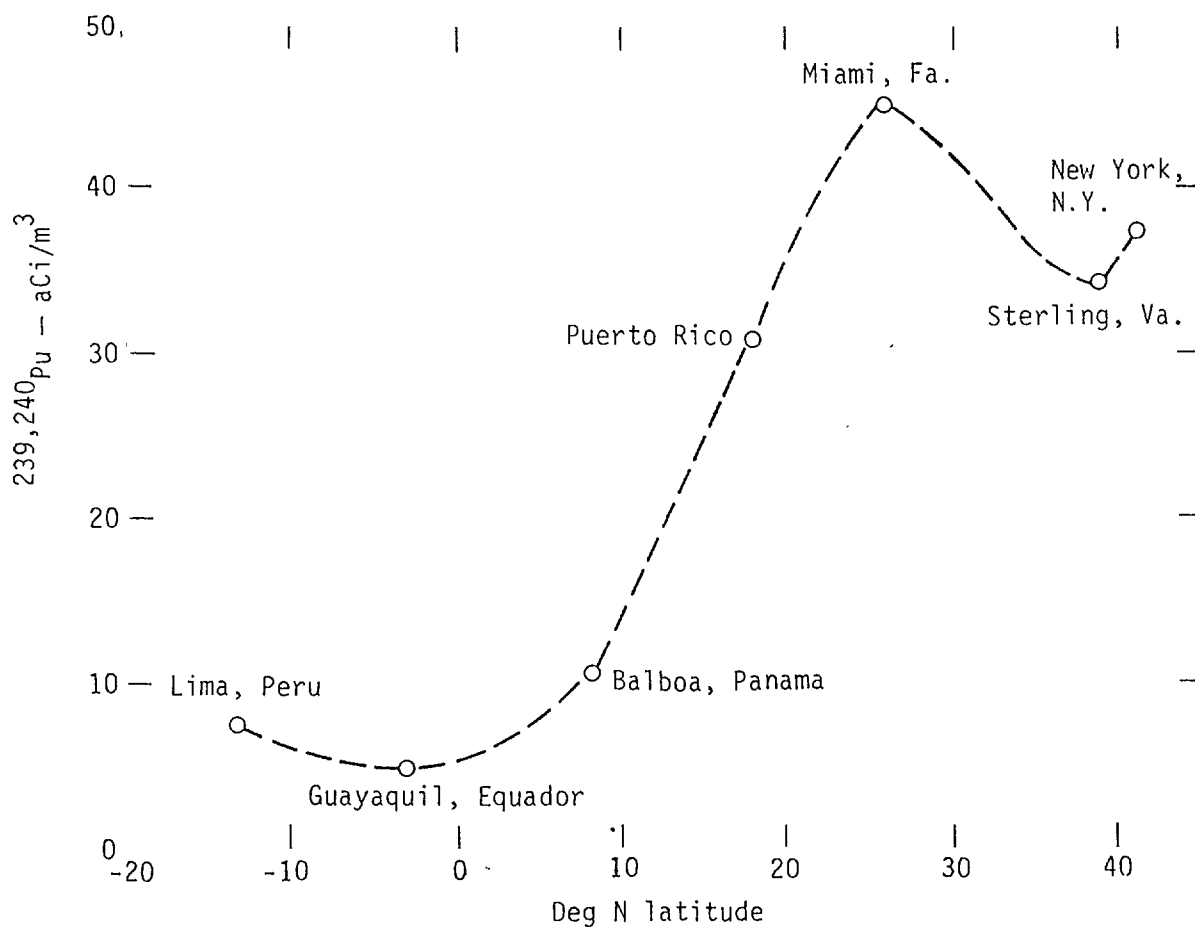


Fig. 2. Concentration of $^{239,240}\text{Pu}$ in air at various latitudes, taken during May 1972, by the HASL sampling network.

Table 1. Radionuclide concentrations in the drinking water of Bikini cisterns, pCi/liter in June 1975.

Building that the cistern collects from	^{137}Cs	^{90}Sr	$^{239,240}\text{Pu}$
5	2.5	1.1	7.9×10^{-3}
24	1.8	1.9	13.7×10^{-3}
School	1.7	1.4	29.0×10^{-3}

drained from the windward roof of each building. Ground water has also been used for drinking in periods of drought and will be used in the future whenever cistern water is unavailable. The ground water is presently in high demand for agriculture on Bikini Island.

Three of the cisterns were first sampled in June 1975 and analyzed for ^{137}Cs , ^{90}Sr , and plutonium radionuclides. The results are abstracted in Table 1.⁹ From an examination of the fallout in rainfall at other Pacific islands over the period 1968 to 1974, it was concluded that the ^{90}Sr and, by analogy, ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in the cistern water did not result solely from worldwide fallout.⁹ Instead, the cisterns appear to have contained locally derived levels of radionuclides. In support of this contention, two water samples collected by one of the authors (V.N.) in October 1975 from the drinking water tanks on the ERDA-supported Marshall Island research vessel, the R. V. Likatanur,

contained 0.6 ± 0.2 fCi/liter of $^{239,240}\text{Pu}$ and 0.09 ± 0.04 pCi/liter of ^{137}Cs . This water came from the rain-water supply collected at Kwajalein Atoll and is therefore indicative of fallout concentrations in Marshall Island rainfall. The $^{239,240}\text{Pu}$ and ^{137}Cs concentrations in Marshall Island rainfall are then approximately 1/20 of the concentrations in the Bikini cisterns. We therefore conclude that the Bikini cistern water contains levels of plutonium radionuclides that, while small, are nevertheless significantly elevated above the levels expected from worldwide fallout. The higher concentrations could originate from leaching of the concrete cisterns (the concrete used for the cistern construction was locally derived) or, alternatively, from labeled particles being resuspended in air, accumulating on the drainage surface of the roofs, then being washed into the cisterns with the next rain. If the latter mechanism is correct, resuspension processes contribute

plutonium not only to the inhalation pathway but to the ingestion pathway as well.

Bennett³ has recently published data on fallout ^{239,240}Pu in 1972 dietary components in New York that included a mean tap water concentration of 0.3 fCi/liter from 1973. Other data appropriate for comparative purposes are fallout levels in untreated surface water of the Great Lakes.^{10,11} These data are summarized in Table 2, along with the mean and range of plutonium concentrations in cistern and ground water from Bikini Island. Assuming that water consumption rates for individuals at Bikini and New York are similar, we see that Bikinians experience a higher plutonium body

burden from cistern or ground water ingestion than do New Yorkers. We assume here, of course, that Bikini Island water is the only available source for the present population. It follows that urine levels in the Bikini population would exceed those in a New York population even if drinking water were the only pathway involved and each of the other pathways contributed a similar level of plutonium at Bikini and New York.

Dietary Intake

Terrestrial Food Pathway — The diet for the people on Bikini Island consists of foods imported from the United States and foods grown on Bikini Atoll. The imported foods,

Table 2. Comparison of plutonium concentrations in water of Bikini Island with those in several U.S. sources.

Location	^{239,240} Pu, fCi/liter		Data from Ref. No.
	Mean	Range	
Bikini (1975)			
Cistern water	17	8-29	9
Ground water	44	6-122	9
New York (1973)			
City tap water	0.3		3
Great Lakes (1973)			
Superior	0.63		10
Michigan	0.73		10
Huron	0.63		10
Erie	0.17		10
Ontario	0.25		11

Table 3. Fallout $^{239,240}\text{Pu}$ in a typical diet for New York, 1972^a; data from Ref. 3.

Food product	Consumption, kg/yr	Concentration, pCi/kg (fresh)	Intake, pCi/yr
Shellfish	1	0.011	0.011
Bakery products	44	0.0085	0.37
Whole grain products	11	0.0060	0.066
Fresh fruit	59	0.0051	0.30
Dry beans	3	0.0048	0.014
Fresh vegetables	48	0.0043	0.21
Root vegetables	10	0.0035	0.035
Poultry	20	0.0033	0.066
Flour	34	0.0028	0.095
Meat	79	0.0026	0.20
Fresh fish	8	0.0016	0.013
Rice	3	0.0016	0.005
Potatoes (peeled)	38	0.0014	0.053
Eggs	15	0.0012	0.019
Macaroni	3	0.0012	0.004
Canned vegetables	22	0.0009	0.019
Milk	200	<0.0003	<0.06
Fruit juice	28	<0.0003	<0.007
Canned fruit	11	<0.0002	<0.002
Tap water (1973)	511	0.0003	0.12
Total intake			1.6

^aExcept for tap water - 1973.

on the average, should contain fallout levels of plutonium similar to those in food consumed by New Yorkers. Plutonium concentrations in the New York diet are abstracted from a report by Bennett in Table 3.³

Some recent plutonium data from the June 1975 Bikini Survey by one of the authors⁴ and earlier data

obtained for terrestrial food items¹² are given in Table 4. Only plutonium detection limits (with 95% confidence) were available for some samples because of the limited sample size obtainable from the existing inventory of food products on the island. However, from those samples (papaya, pig muscle) where there was sufficient

material to obtain a real number, it is clear that the plutonium concentrations are almost 100 times any shown in Table 3 for terrestrially derived food products in the New York diet.

It is not yet clear just how much of the different food products grown on Bikini Island are actually used in the diet. However, whatever the use (and there is undoubtedly some) and whatever increased future use

Table 4. Concentrations of $^{239,240}\text{Pu}$ in foods grown on Bikini Island^a; the sign "<" denotes the detection limit for that sample; actual concentration was somewhere below this limit.

Food product	Concentration, pCi/kg (fresh)
Bikini June 1975	
Pandanus	<2.7
Breadfruit	<3.6
Papaya	0.67
Coconut	<0.27
Squash	<3.6
Pig muscle	0.72
Chicken flesh	<6.3
Bikini 1974	
Banana ^b	<3.6
Papaya ^b	<1.8
Squash ^b	<9.0
Pandanus ^b	<1.8

^aData from a survey by one of the authors.⁴

^bData from Ref. 12.

there may be, use of food products from Bikini would lead to higher body burdens and therefore higher concentrations of plutonium in the urine of the Bikini population.

For example, if one assumes an average plutonium concentration of 0.6 pCi/kg fresh weight in all the terrestrial food products of Bikini Island and a combined intake of all foods of 100 g/day or 36.5 kg/yr, then the yearly plutonium intake would be 21.9 pCi, compared to the 1.45 pCi (1.6 pCi minus the intake from shellfish, fresh fish, and water) estimated by Bennett³ for New York. Thus, plutonium levels in a diet entirely derived from Bikini terrestrial foods are 15 times the levels in a terrestrial diet from New York.

Marine Food Pathway — Marine food products from Bikini Atoll supply a substantial amount of plutonium to the diet for the inhabitants. Some plutonium concentrations in fish at Bikini Atoll have been published by Nevissi and Schell.¹³ These data are abstracted in Table 5 along with some data on concentrations in marine invertebrates.¹⁴ Using the concentrations in fish muscle and eviscerated whole fish and the detection limits (denoted in Table 5 by "<") as real numbers, and weighting by the number of fish per sample, we find the mean concentration of plutonium

Table 5. Concentrations of $^{239,240}\text{Pu}$ in Bikini fish (combined data for fish muscle and eviscerated whole fish) and invertebrates; the sign "<" denotes the detection limit for that sample; actual concentration was somewhere below this limit.

Species	No. of fish per sample	$^{239,240}\text{Pu}$ pCi/kg wet	Data from Ref. No.
Surgeon fish	3	<0.45	13
Surgeon fish	1	8.1	13
Convict surgeon	39	<0.45	13
Convict surgeon	4	12.6	13
Convict surgeon	1	4.5	13
Convict surgeon	4	7.7	13
Panulirus (lobster)	8	<0.4	14
Grapsus (crab)	5	1.7 ± 0.5	14

in fish at Bikini Atoll is 2.2 pCi/kg wet weight. Concentrations in invertebrate muscle average 1 pCi/kg. If we assume an average daily intake of 600 g (or 219 kg/yr),¹⁵ the total annual plutonium intake would be 482 pCi.

For comparison, the data listed in Table 3 show a concentration in

New York shellfish and fish of 0.011 pCi/kg and 0.0016 pCi/kg wet weight, respectively. The total annual intake of plutonium from New York marine products would be 0.024 pCi. Thus, the plutonium intake through the marine food pathway in Bikini is 2×10^4 the intake through the same pathway in New York.

Intake Concentrations Compared with Excreted Concentrations

The estimated annual intake of plutonium through various pathways is given in Table 6. With the possible exception of the inhalation pathway, available data indicate that all exposure pathways will contribute a higher plutonium body burden to Bikinians than to New York residents.

Since surface soil concentrations are much higher at Bikini than in New York, and the material resuspended by a person in his immediate environment may be more important than open-air plutonium concentrations for estimating intake by inhalation, we believe that the plutonium intake via

inhalation at Bikini Island also exceeds that in New York.

An inhalation pathway analysis based on a mass-loading concept similar to the one used for the Enewetak Atoll dose assessment¹⁵ also indicates potentially higher intake at Bikini by inhalation. Using a mass loading of $100 \mu\text{g}/\text{m}^3$ as in the Enewetak assessment; an average, 0- to 5-cm concentration of plutonium in Bikini soil of approximately $9 \text{ pCi}/\text{g}$;⁴ and a breathing rate of $20 \text{ m}^3/\text{day}$; we find the annual intake via inhalation would be 6.6 pCi , compared with the estimate of 0.2 pCi based on aerosol measurements.

Inhalation experiments¹⁶ and dose models^{3,16,17} indicate that approximately 0.1% of the activity inhaled will be excreted in the urine. If we assume that a person's annual intake via inhalation is 0.2 pCi in both Bikini and New York, inhalation would contribute only $2 \times 10^{-4} \text{ pCi}$

of plutonium to a person's urine during the course of a year.

The transfer coefficient across the gut into the blood is assumed to be 3×10^{-5} for ingested plutonium.¹⁶ At Bikini this would mean that $1.54 \times 10^{-2} \text{ pCi}$ would be transferred to the blood. Of this amount, approximately 8%^{16,17} or $1.2 \times 10^{-3} \text{ pCi}$ would be transferred to the urine. We find from this analysis that at Bikini the ingestion pathway contributes more than the inhalation pathway to plutonium in the urine of man.

In New York, the contribution of ingestion to the total level of plutonium in urine ($1.6 \text{ pCi} \times 3 \times 10^{-5} \times 0.08 = 3.8 \times 10^{-6} \text{ pCi}$) is negligible compared to the contribution of the inhalation route. Therefore the estimated total annual plutonium in urine would be $2 \times 10^{-4} \text{ pCi}$ for New York residents and $2 \times 10^{-4} + 1.2 \times 10^{-3} = 1.4 \times 10^{-3} \text{ pCi}$ for

Table 6. Comparison of estimated annual intakes of plutonium at Bikini with those at New York.

Pathway	Est. annual intake, pCi/yr Bikini	New York	Ratio of intakes, Bikini/New York
Inhalation	≥ 0.2	0.2	≥ 1
Drinking water ^a	8.7	0.13	58
Terrestrial foods	21.9	1.4	15
Marine foods	482	0.024	2×10^4

^aCistern water only. Any use of ground water would increase this estimate.

Table 7. Concentration of Pu in urine — measured vs predicted.

Location	Estimated annual intake, pCi		Predicted annual Pu in urine; computed from estimated annual intake ^a			Annual quantity of Pu excreted in urine, ^b pCi
	Inhaled	Ingested	Total pCi	% from ingestion	% from inhalation	
New York	0.2	1.6	0.0002	3	97	0.365
Bikini	0.2	513 ^c	0.0014	86	14	3.65

^aModel and parameters are those summarized in Refs. 3 and 17.

^bComputed from assumed excretion rate of 1 liter/day and concentrations reported by Ref. 2.

^cSum of estimated Bikini intakes for drinking water, terrestrial foods and marine foods from Table 6.

Bikini residents. This computation shows that plutonium levels in the urine should presently be at least seven times higher at Bikini than at New York. This ratio is very similar to that reported to us for plutonium in recent urine samples: a Bikini/New York ratio of about ten.²

These analyses indicate that the differences observed in the plutonium concentrations in urine of New York and Bikini populations can be accounted for by the estimated differences of plutonium intake via food, water, and air, based on the assumed dietary and inhalation intakes. However, the absolute quantities of excreted plutonium predicted by models^{16,17} based on the same dietary intake values (given in Table 6), do not agree with the urine concentrations recently reported to us.² Table 7 compares quantities computed from the reported urine concentrations with the quantities predicted from the pathways and models for New York and Bikini.

The reported concentration of plutonium in urine from the New York population is 10^{-3} pCi/liter.² If we assume a urine excretion of 1 liter/day per person, the total plutonium excreted via urine per year would then be 0.365 pCi. (See Table 7.) Bennett estimates the total annual intake of plutonium via food, water, and inhalation to be approximately 1.8 pCi for a person in New York.³ These values then suggest that 20% of the intake is appearing in the urine. This is a much higher percentage than has ever been reported¹⁶ and is higher than percentages normally used for model predictions.^{3,17}

Only 8% of the amount of plutonium entering the blood reaches the urine,¹⁷ while 90% of the plutonium entering the blood is equally partitioned between the liver and the bones.^{3,16,17} Thus, if 0.365 pCi (the total plutonium excreted in urine per year, based on the concentrations reported for New York) is 8% of the plutonium in the blood,

and if the bones and the liver each receive 45% of that in the blood, then the bone and liver burdens of New York residents should each increase by 2.1 pCi annually. However, a 4.2 pCi increase is nearly twice the present New York total body burden accumulated since 1954.⁸ The plutonium concentrations reported for the urine of the Bikini population would, of course, indicate body burdens ten times higher than those of the New York population.

Bennett's data³ can also be used to predict the quantity of plutonium expected in the urine as a result of the body burden accumulated since 1954. The major source of input from the accumulated body burden to the blood, and subsequently to the urine, is from turnover in the lungs and lymph nodes, which have half times of 500 and 1000 days, respectively. Bennett's data show an accumulated plutonium burden in the lymph-nodes of 0.40 pCi and in the lungs of 0.12 pCi for a typical New Yorker in the year 1974. The plutonium appearing in the urine from these two compartments during the next year would be 0.013 pCi. The contribution to the urine from inhalation and ingestion of plutonium during the current year would be 0.0002 pCi. (See Table 7.) Therefore, the expected annual excretion from previous body burdens and present ingestion pathways

is approximately 0.013 pCi, compared with the annual amount of 0.365 pCi computed from the urine concentrations reported by Ref. 2. These results suggest several possible interpretations:

- The urine samples were contaminated at the time of collection, and the plutonium concentrations are actually significantly below those reported.
- The transfer coefficient across the gut for biologically complexed plutonium is much higher than 3×10^{-5} , the value developed from animal studies with various plutonium compounds.
- The transfer to urine from the blood for ingested plutonium is greater than 8%.
- The direct transfer of inhaled plutonium to the blood from the upper respiratory tract is greater than 1% (Ref. 17).
- The estimated intake values through food, water, and air are incorrect.
- Any combination of the above.

Interestingly, the plutonium body distribution models^{3,16,17} and the Bikini pathway data (Tables 6 and 7) show that the major fraction of plutonium presently entering the urine is by ingestion. The absolute quantity of plutonium predicted to reach the urine as a result of this annual intake is 1.4×10^{-3} pCi,

instead of the annual 3.65 pCi computed from the reported data.² There would, of course, be an additional contribution to urine plutonium levels due to transport from deep-lung and lymph-node burdens of previous inhalation exposure. However, the magnitude of this contribution should be similar to that previously estimated for New York and would therefore lead to total annual urine levels of approximately 0.015 pCi. The reported urine concentrations indicate an annual urine level of 3.65 pCi. If we accept these reported urine concentrations for Bikini, then it would seem that the transfer coefficient across the gut for plutonium incorporated in food products must be much higher than 3×10^{-5} .

However, the New York data indicate that the major contribution to the plutonium urine concentrations is via

the inhalation pathway. If the plutonium concentrations reported for urine samples from New York are accepted, the data suggest that the parameters for transfer of inhaled plutonium to blood must be considerably higher than those presently assumed. However, increases in both the percentage transferred from the upper respiratory tract to the blood and in the percentage transferred from the blood to the urine that are reasonable for physiological function and chemical transport still cannot account for 20% of the total annual intake appearing in the urine. Because of this seemingly large fraction of the total intake appearing in the urine, it is difficult to evaluate if this human data indicates that transfer to urine is greater for human populations than previously assumed from animal studies.

Summary

Bikini Atoll may be the only global source of data on humans where intake via ingestion is thought to contribute the major fraction of plutonium body burden. It is possibly the best available source of data for evaluating the transfer of plutonium across the gut wall after being incorporated into biological systems. If the plutonium urine data for the

Bikini population is correct, and the estimated dietary intakes are reasonable, it appears that the transfer across the gut wall of plutonium incorporated into food products is greater than previously expected.

The New York data, where the major intake is via the inhalation pathway, if accepted as reported also lead to altered conclusions regarding the

physiological transport of plutonium. The reported levels in urine would account for as much as 20% of the total estimated annual intake of plutonium. This is a fraction that is much higher than believed possible. If, however, the estimated annual

intake of plutonium and the reported urine concentrations for New York are correct, then plutonium is eliminated more rapidly through the urine than previously estimated. This would indicate that smaller body burdens would be expected from intake of plutonium.

References

1. R. A. Conard, *A Twenty-Year Review of Medical Findings in a Marshallese Population Accidentally Exposed to Radioactive Fallout*, Brookhaven National Laboratory, Brookhaven, Conn., Rept. BNL-50424 (1975).
2. T. F. McCraw, USERDA Headquarters, private communications (1976).
3. B. G. Bennett, "Transuranic Element Pathways to Man," in *Int. Symp. Transuranium Nuclides Environ.*, San Francisco, 1975 (Int. At. Energy Agency, Vienna, 1976), p. 367.
4. C. S. Colsher, W. L. Robison, and P. H. Gudiksen, *Evaluation of the Radionuclide Concentrations in Soil and Plants from the 1975 Terrestrial Survey of Bikini and Eneu Islands*, Lawrence Livermore Laboratory, Rept. UCRL-51879, part 3 (in publication).
5. E. P. Hardy, P. W. Krey, and H. L. Volchok, *Nature* 241, 444 (1973).
6. A. E. Smith and W. E. Moore, *Report of the Radiological Clean-Up of Bikini Atoll*, Southwestern Radiological Health Laboratory, Rept. SWRHL-111r (1972).
7. O. O. T. Lynch, T. F. McCraw, V. A. Nelson, and W. E. Moore, *Radiological Resurvey of Food, Soil, and Groundwater at Bikini Atoll, 1972*, U.S. Energy Research and Development Administration, Rept. ERDA-34 (1975).
8. *Health and Safety Laboratory Fallout Program, Quarterly Environmental Appendix*, Health and Safety Laboratory, N.Y., Rept. HASL-306, Append. (July, 1976).
9. V. E. Noshkin, K. M. Wong, R. J. Eagle, and G. Brown, *Preliminary Evaluation of the Radiological Quality of the Water on Bikini and Eneu Islands*, Lawrence Livermore Laboratory, Rept. UCRL-51971 (1975).
10. M. A. Wallgren and D. M. Nelson, *Plutonium in the Five Great Lakes; Comparison of Surface Waters*, Argonne National Laboratory, annual Rept. ANL-8060 Part III (1973), p. 93.
11. J. G. Farmer, V. T. Bowen, and V. E. Noshkin, *Long Lived Artificial Radionuclides in Lake Ontario, Part I. Supply from Fallout, and Concentration in Lake Water of Plutonium, Americium, Strontium 90 and Cesium 137*, Woods Hole Oceanographic Institute, Woods Hole, Mass., Rept. COO3568-6 (1976).
12. V. Nelson, University of Washington, private communication (Nov. 1976).
13. A. Nevissi and W. R. Schell, *Nature* 255, 321 (1975).
14. V. Noshkin, unpublished data.

15. *Enewetak Radiological Survey*, U.S. Atomic Energy Commission, Nevada Operations Office, Las Vegas, Nevada, Rept. NVO-140 vol. 1 (1973).
16. *The Metabolism of Compounds of Plutonium and Other Actinides*, International Commission on Radiological Protection, Publ. 19 (1972).
17. W. E. Martin and S. G. Bloom, "Plutonium Transport and Dose Estimation Model," in *Int. Symp. Transuranium Nuclides Environ., San Francisco, 1975* (Int. At. Energy Agency, Vienna, 1976), p. 385.

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